

Thermodynamic Excess Properties and FTIR Spectroscopic Studies of Binary Mixtures Containing Propiophenone and 2-Alkoxy Ethanol at Temperatures 303.15K, 308.15K, 313.5K and 318.15K

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Abstract: Densities ρ , viscosities η and ultrasonic speeds of sound of binary mixtures of Propiophenone (PPH) with 2-Alkoxy Ethanol [2-Methoxy Ethanol, 2-Ethoxy Ethanol, 2-Butoxy Ethanol] have been measured over the entire composition range from 303.15K to 318.15K and at atmospheric pressure 0.1MPa. The worked out experimental data has been used to calculate various acoustical parameters like excess molar volume V^E , excess ultrasonic velocity U^E , acoustic impedance Z , excess acoustic impedance (Z^E), isentropic compressibility K_S , excess isentropic compressibility K_S^E , intermolecular free length L_f , excess intermolecular free-length (L_f^E), deviation in viscosity $\Delta\eta$ were calculated and the computed results were fitted with the Redlich Kister equation to estimate the binary coefficients and standard deviation between experimental and calculated data. At different concentrations, FTIR analysis has been done for the above binary mixtures to confirm the hydrogen bonding presence.

Keywords: Ultrasonic speed, impedance, excess molecular volume, Intermolecular free-length, Viscosity, isentropic compressibility.

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I. Introduction

Propiophenone is a generally used component in some perfumes. Propiophenone is used in the synthesis of ketoamphetamines such as cathinone and methcathinone. Propiophenone can also be converted to synthetic aryl alkenes such as cinnamic acids. Properties such as density, viscosity of pure chemicals and of their binary mixtures over the whole composition range at a particular temperature or several temperatures, are useful for a complete understanding of their thermodynamic properties and for practical chemical engineering purposes. Physico chemical and thermodynamic investigations play an important role to understand the nature and extent of the aggregation of molecules that exists in the binary liquid mixtures and their sensitivities to the variations in composition and the molecular structure of the pure components [1, 2]. In the recent years there has been considerable interest in theoretical and experimental investigations of the excess thermodynamic properties of binary mixtures.

Density and ultrasonic velocity properties of binary liquid mixtures are essential in process simulation, equipment design, solution theory, important parameter in calculations of the thermodynamic properties of matter and molecular dynamics. The ultrasonic velocity measurement is an excellent tool to investigate inter and intra molecular interactions between liquid mixtures of non-electrolytes. Acoustic properties are useful in understanding the molecular interactions.

The present research paper presents the data on densities, viscosity and ultrasonic velocity of binary liquid mixtures of Propiophenone (PPH) with 2-Alkoxy Ethanol [2-Methoxy Ethanol (2-MOE), 2-Ethoxy Ethanol (2-EOE), 2-Butoxy Ethanol (2-BOE)] at temperatures 303.15K, 308.15K, 313.K, and 318.15K. By using this data, various acoustical parameters like Acoustic impedance (Z), Isentropic compressibility (K_S), Intermolecular free-length (L_f) and also various excess properties like excess ultrasonic velocity (V^E), excess acoustic impedance (Z^E), excess isentropic compressibility (K_S^E) and excess intermolecular free-length (L_f^E) were calculated and fitted to the Redlich Kister equation to estimate the standard deviations. Above results are used to explain the nature of intermolecular interactions between mixing components. This work provides a test of

various empirical equations to correlate viscosity and acoustic data of binary mixtures in terms of pure component properties.

II. Material and methods

Propiophenone, 2-Alkoxy Ethanol [2-Methoxy Ethanol (2-MOE), 2-Ethoxy Ethanol (2-EOE), 2-Butoxy Ethanol (2-BOE)], these chemicals purchased from S.D. Fine chemicals Ltd, India and used in the present investigation, details as shown in Table-01a.

Table-01a: Provenance and purity of the materials used

Provenance and purity of the materials used					
CHEMICALS	CAS number	Source	Water content (%)	Mass fraction purity(final)	
Propiophenone	93-55-0	S.D fine Chemicals, India	0.00049	> 98%	
2-Methoxy ethanol	109-86-4	S.D fine Chemicals, India	0.0004	0.993	
2-Ethoxy ethanol	110-80-5	S.D fine Chemicals, India	0.0004	0.987	
2-Butoxy ethanol	111-76-2	S.D fine Chemicals, India	0.0004	0.994	

Mixtures were prepared by mixing weighed amounts of the pure liquids adopting the method of closed system by using Mettler Toledo (ME204)balance with the precision of ± 0.1 mg. Mixtures were allowed to stand for some time before every measurement so as to avoid air bubbles. The purities of the liquids were checked by comparing the values of densities and ultrasonic velocities with literature data and are given in Table-01b.

Table-01b: Physical Properties Of Pure Components

Comparison of experimental values of densities, ultrasonic velocity and Viscosities of pure liquids with the corresponding literature values at various temperatures

Compound	T(K)	Density (ρ)		Ultrasonic speed(u)		Viscosity(η)	
		Experimental	literature	Experimental	literature	Experimental	literature
		gm/cm ³	gm/cm ³	m. s ⁻¹	m. s ⁻¹	m.pas	m.pas
Propiophenone	303.15	1.0045		1446.41		1.515	
	308.15	1.0015	1.00896 ^a 1.0087 ^b	1427.038		1.469	1.469 ^a 1.468 ^c
	313.15	0.9985		1403.95		1.423	
	318.15	0.9955		1380		1.377	
2-Methoxy Ethanol	303.15	0.9525	0.9527 ^a	1347.52	1324.3 ^f	1.57265	1.544 ^g
	308.15	0.9492	0.95110 ⁱ	1340.12		1.39211	
	313.15	0.9456	0.94629 ^j	1333.79		1.22566	
	318.15	0.9421	0.94162 ^j	1327.36		1.095	1.0789 ^h
2-EthoxyEthanol	303.15	0.92117	0.9204 ^f	1318.24	1319.2 ^d	1.57219	1.6315 ^g
	308.15	0.91643	0.91640 ^k	1296.572		1.46924	1.480 ^k
	313.15	0.91227	0.91229 ^j	1268.646		1.36754	1.293 ^l
	318.15	0.90784	0.90790 ^k	1235	1235.1 ^e	1.2785	1.2309 ^h
2-ButoxyEthanol	303.15	0.89237	0.8921 ^a	1288	1288.02 ^d	2.4279	2.4031 ^g
	308.15	0.88771		1272	1275 ^e	2.27854	
	313.15	0.88326		1259	1259 ^e	2.12115	
	318.15	0.87939		1242	1242.1 ^e	1.92765	

a[3], b[4], c[5], d[6], e[7], f[8], g[9], h[10], i[11], j[12], k[13], l[14]

Proper care was taken to avoid any evaporation loss while doing the experiment. The densities of liquids and their mixtures were measured using bi-capillary pycnometer having a capillary diameter of 0.85 mm, which was calibrated using double distilled water. The necessary buoyancy corrections were applied. The density (ρ) values were reproducible within ± 0.2 Kg m⁻³. The ultrasonic velocity (u) measurements were made by a single frequency (2 MHz) variable path interferometer with an error of ± 0.0001% ultrasonic interferometer model F-05(s.no.1415342) Mittal Enterprises, New Delhi. Formulae used are:

$$V^E = \frac{x_1M_1 + x_2M_2}{\rho_m} - \left\{ \frac{x_1M_1}{\rho_1} + \frac{x_2M_2}{\rho_2} \right\} \dots\dots\dots (1)$$

$$K_S = U^{-2} \rho^{-1} \dots\dots\dots (2)$$

$$K_S^E = K_S - K_S^{iD} \dots\dots\dots (3)$$

$$K_S^{iD} = \sum_{i=1}^2 \phi_i \left(K_{Si} + \frac{TV_i\alpha_i^2}{C_{pi}} \right) - \frac{T(\sum_{i=1}^2 x_i V_i)(\sum_{i=1}^2 \phi_i \alpha_i)}{\sum_{i=1}^2 x_i C_{pi}} \dots\dots (4)$$

$$\Delta u = u - (x_1u_1 + x_2u_2) \dots\dots\dots (5)$$

$$\Delta \eta = \eta - (x_1\eta_1 + x_2\eta_2) \dots\dots\dots (6)$$

In the above equations: M_i , K_S^E , η_i , u_i and ρ_i represent the molecular weight, isentropic compressibility, viscosity, ultrasonic velocity and density of component.

Where ϕ_i is the ideal state volume fraction and is defined by the relation:

$$\phi_i = \frac{x_i V_i}{\sum_{i=1}^2 x_i V_i} \dots \dots \dots (7)$$

III. Results and Discussions:

The experimental values of Ultrasonic velocity, density and viscosity of three binary mixtures at temperatures 303.15, 308.15, 313.15 and 318.15 K are presented in Table-02. From these values, it is observed that, in all the systems, ultrasonic speed (u) and density (ρ) increased non-linearly with mole fraction of PPH and the viscosity is decreased at 303.15k for all three binary mixtures, where as in the case of other temperatures, the viscosity is increased for the binary mixtures of PPH+2-MOE & PPH+2-EOE, but in the case of PPH+2-BOE, viscosity is decreased at all temperatures.

Table-02:Experimental values at temperatures T=303.15, 308.15, 313.15 and 318.15 K
 Densities (ρ /g·cm⁻³), Ultrasonic velocities (u/m·sec⁻¹), and viscosities (η /mPa.s) of binary mixtures of Propiophenone with 2-Alkoxy Ethanol at T=303.15, 308.15, 313.15 and 318.15 K

X1	303.15K			308.15K			313.15K			318.15K		
	ρ	U	η	ρ	U	η	ρ	U	η	ρ	U	η
	g cm-3	m.s-1	m. pas	g cm-3	m.s-1	m. pas	g cm-3	m.s-1	m. pas	g cm-3	m.s-1	m. pas
PPH+2-MOE												
0.0000	0.9525	1347.52	1.5727	0.9492	1340.12	1.3921	0.9456	1333.79	1.2257	0.9421	1327.36	1.0950
0.0624	0.9574	1357.24	1.5676	0.9542	1348.10	1.3945	0.9507	1340.06	1.2344	0.9474	1332.19	1.1078
0.1302	0.9622	1367.21	1.5623	0.9592	1356.18	1.3983	0.9558	1346.66	1.2463	0.9525	1337.39	1.1251
0.2041	0.9671	1376.87	1.5567	0.9642	1364.68	1.4025	0.9610	1353.86	1.2593	0.9578	1343.04	1.1447
0.2852	0.9721	1386.35	1.5509	0.9693	1373.70	1.4076	0.9662	1361.17	1.2743	0.9630	1348.33	1.1666
0.3744	0.9772	1395.90	1.5452	0.9745	1382.35	1.4140	0.9714	1368.04	1.2917	0.9682	1353.24	1.1916
0.4731	0.9824	1405.21	1.5398	0.9797	1390.80	1.4216	0.9767	1374.83	1.3116	0.9735	1358.05	1.2197
0.5827	0.9878	1414.36	1.5342	0.9850	1399.00	1.4310	0.9820	1381.36	1.3339	0.9790	1362.71	1.2514
0.7054	0.9932	1423.89	1.5290	0.9904	1407.39	1.4419	0.9875	1388.08	1.3593	0.9845	1367.50	1.2870
0.8434	0.9988	1434.44	1.5228	0.9959	1415.96	1.4545	0.9930	1395.33	1.3883	0.9900	1373.05	1.3280
1.0000	1.0045	1446.41	1.5150	1.0015	1427.04	1.4690	0.9985	1403.95	1.4230	0.9955	1380.00	1.3770
PPH+2-EOE												
0.0000	0.9212	1318.24	1.5722	0.9164	1296.57	1.4692	0.9123	1268.65	1.3675	0.9078	1235.00	1.2785
0.0756	0.9290	1330.96	1.5623	0.9246	1318.17	1.4655	0.9206	1280.48	1.3665	0.9165	1247.12	1.2791
0.1554	0.9369	1344.01	1.5547	0.9328	1330.31	1.4636	0.9291	1293.74	1.3688	0.9250	1260.68	1.2846
0.2397	0.9450	1357.37	1.5471	0.9411	1342.62	1.4621	0.9375	1306.90	1.3721	0.9337	1274.51	1.2915
0.3291	0.9532	1370.23	1.5396	0.9494	1354.35	1.4612	0.9460	1319.95	1.3763	0.9422	1288.46	1.3001
0.4239	0.9615	1382.81	1.5324	0.9578	1366.09	1.4609	0.9546	1332.93	1.3813	0.9509	1302.44	1.3094
0.5246	0.9699	1395.00	1.5264	0.9664	1377.32	1.4613	0.9632	1346.01	1.3873	0.9597	1316.61	1.3198
0.6319	0.9784	1407.16	1.5217	0.9750	1388.49	1.4623	0.9720	1359.33	1.3941	0.9685	1331.09	1.3310
0.7464	0.9870	1419.18	1.5175	0.9838	1400.13	1.4635	0.9808	1372.71	1.4014	0.9775	1345.75	1.3431
0.8688	0.9957	1432.21	1.5149	0.9927	1412.93	1.4655	0.9896	1387.56	1.4102	0.9865	1361.86	1.3572
1.0000	1.0045	1446.41	1.5150	1.0015	1427.04	1.4690	0.9985	1403.95	1.4230	0.9955	1380.00	1.3770
PPH+2-BOE												
0.0000	0.8924	1288.00	2.4279	0.8877	1272.00	2.2785	0.8833	1259.00	2.1212	0.8794	1242.00	1.9277
0.0996	0.9030	1306.54	2.3328	0.8987	1289.80	2.1924	0.8945	1275.29	2.0439	0.8908	1257.06	1.8626
0.1994	0.9139	1325.16	2.2401	0.9098	1306.97	2.1095	0.9058	1291.36	1.9720	0.9023	1272.23	1.8053
0.2992	0.9249	1342.81	2.1477	0.9210	1324.03	2.0271	0.9172	1307.17	1.9010	0.9137	1287.41	1.7490
0.3990	0.9360	1359.52	2.0558	0.9322	1340.50	1.9457	0.9286	1322.62	1.8311	0.9252	1301.93	1.6938
0.4990	0.9472	1375.28	1.9643	0.9436	1356.30	1.8647	0.9402	1337.41	1.7615	0.9368	1315.87	1.6390
0.5990	0.9585	1390.12	1.8731	0.9550	1371.26	1.7841	0.9517	1351.24	1.6919	0.9484	1329.19	1.5842
0.6992	0.9698	1404.46	1.7824	0.9665	1385.38	1.7041	0.9633	1364.61	1.6223	0.9601	1341.84	1.5296
0.7994	0.9812	1418.45	1.6920	0.9781	1399.14	1.6244	0.9749	1377.48	1.5540	0.9718	1354.23	1.4761
0.8996	0.9928	1432.28	1.6024	0.9898	1412.79	1.5452	0.9867	1390.18	1.4861	0.9837	1366.54	1.4237
1.0000	1.0045	1446.41	1.5150	1.0015	1427.04	1.4690	0.9985	1403.95	1.4230	0.9955	1380.00	1.3770

This non-linear variation is a deviation from ideal behavior which suggests the presence of intermolecular interactions between the component molecules of the mixtures, however the strength of interaction is well reflected in the excess parameters like excess molar volume (V^E), and deviation in viscosity ($\Delta\eta$) etc., as these parameters are found to be more sensitive towards intermolecular interactions in the liquid mixtures [15].Larger dispersionwith temperature at lower values ofmole fractions of PPH has been observed. However this dispersion gradually decreased with mole fraction of PPH and is almost zero at X1=1. The dominance of PPH over oxy-ethanol is in breaking the intramolecular associations; followed by new hydrogen bond formation (oxygen atom of Propiophenone and the hydrogen atom of alkoxyethanol) can be inferred.

This is further supported by decrease of K_S and L_f values in MOE, EOE and BOE; From the thermodynamic point of view — the ultrasonic velocity, isentropic compressibility, deviation in isentropiccompressibility, free length along with the other data like excess molar volume, viscosity, are strongly affected by the changes of concentration and temperature, besides the type of bonding present between the

molecules of the constituent liquids. Hydrogen bonding causes considerable influence on these parameters, because of the fact that specific interactions between the molecules are controlled mainly by hydrogen bond that binds the molecules together and therefore the density variation with temperature is less at higher values of X_1 . Computed values of the parameters analyzed/and their excess values are given in Table 03a to Table 03d. TABLE-03a-3d- Values of excess molar Volume (V_m^E), ultrasonic velocity (Δu), excess isentropic compressibility (Δk_s), excess acoustic impedance (Δz), excess intermolecular free-length (L_f^E) and excess viscosity ($\Delta \eta$) for the binary liquid -mixtures of Propiophenone with 2-MOE, 2-EOE & 2-BOE.

TABLE-03a- At T=303.15k

X1	V^E ($\text{cm}^3 \cdot \text{mol}^{-1}$)	Δu ($\text{m} \cdot \text{s}^{-1}$)	Δk_s (pa^{-1})	Δz ($10^{-3}/\text{Kg} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$)	L_f^E (10-10m)	$\Delta \eta$ ($\text{m} \cdot \text{pas}$)
PPH+2-MOE 303.15K						
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0624	0.0297	3.5551	-0.4766	0.0053	-0.1494	-0.0014
0.1302	0.0627	6.8175	-0.8882	0.0100	-0.2784	-0.0028
0.2041	0.0902	9.1635	-1.1884	0.0135	-0.3726	-0.0042
0.2852	0.1157	10.6291	-1.3779	0.0159	-0.4320	-0.0053
0.3744	0.1323	11.3580	-1.4698	0.0172	-0.4608	-0.0058
0.4731	0.1392	10.9126	-1.4278	0.0168	-0.4476	-0.0056
0.5827	0.1302	9.2164	-1.2454	0.0148	-0.3904	-0.0049
0.7054	0.1034	6.6160	-0.9410	0.0112	-0.2950	-0.0030
0.8434	0.0598	3.5133	-0.5300	0.0064	-0.1661	-0.0012
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
PPH+2-EOE						
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0756	0.0610	3.0380	-0.5786	0.0041	-0.1814	-0.0027
0.1554	0.1051	5.8593	-1.0720	0.0079	-0.3361	-0.0042
0.2397	0.1382	8.4016	-1.4683	0.0112	-0.4603	-0.0056
0.3291	0.1571	9.8143	-1.6968	0.0133	-0.5319	-0.0067
0.4239	0.1651	10.2442	-1.7712	0.0142	-0.5553	-0.0071
0.5246	0.1620	9.5154	-1.6796	0.0135	-0.5265	-0.0070
0.6319	0.1437	7.9307	-1.4472	0.0117	-0.4537	-0.0061
0.7464	0.1137	5.2732	-1.0547	0.0083	-0.3307	-0.0048
0.8688	0.0668	2.6198	-0.5753	0.0044	-0.1804	-0.0029
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
PPH+2-BOE						
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0996	0.0867	2.7567	-0.6898	0.0002	-0.2163	-0.0041
0.1994	0.1396	5.5818	-1.2599	0.0012	-0.3950	-0.0058
0.2992	0.1748	7.4188	-1.6150	0.0018	-0.5063	-0.0071
0.3990	0.1896	8.3045	-1.7800	0.0020	-0.5580	-0.0078
0.4990	0.1925	8.2296	-1.7683	0.0018	-0.5544	-0.0081
0.5990	0.1805	7.2299	-1.6000	0.0012	-0.5016	-0.0080
0.6992	0.1601	5.7031	-1.3155	0.0005	-0.4124	-0.0073
0.7994	0.1255	3.8195	-0.9379	-0.0002	-0.2940	-0.0061
0.8996	0.0754	1.7640	-0.4867	-0.0005	-0.1526	-0.0042
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000

TABLE-03b-At T=308.15K

X1	V^E ($\text{cm}^3 \cdot \text{mol}^{-1}$)	Δu ($\text{m} \cdot \text{s}^{-1}$)	Δk_s (pa^{-1})	Δz ($10^{-3}/\text{Kg} \cdot \text{m}^{-2} \cdot \text{s}^{-1}$)	L_f^E (10-10m)	$\Delta \eta$ ($\text{m} \cdot \text{pas}$)
PPH+2-MOE 308.15K						
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0624	0.0204	2.5589	-0.3955	0.0045	-0.1240	-0.0024
0.1302	0.0458	4.7490	-0.7226	0.0083	-0.2265	-0.0038
0.2041	0.0623	6.8169	-1.0086	0.0117	-0.3162	-0.0053
0.2852	0.0783	8.7865	-1.2454	0.0147	-0.3904	-0.0064
0.3744	0.0893	9.6873	-1.3544	0.0162	-0.4246	-0.0069
0.4731	0.0965	9.5604	-1.3365	0.0162	-0.4190	-0.0068
0.5827	0.0915	8.2296	-1.1788	0.0144	-0.3696	-0.0060
0.7054	0.0750	5.9611	-0.8944	0.0110	-0.2804	-0.0044
0.8434	0.0430	2.5338	-0.4592	0.0056	-0.1440	-0.0025
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
PPH+2-EOE						
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0756	0.0406	2.5624	-0.5878	0.0038	-0.1843	-0.0037
0.1554	0.0796	5.2354	-1.1066	0.0075	-0.3469	-0.0056
0.2397	0.1069	7.5028	-1.5101	0.0107	-0.4734	-0.0071
0.3291	0.1295	8.5531	-1.7231	0.0123	-0.5402	-0.0080
0.4239	0.1411	8.7809	-1.7871	0.0129	-0.5603	-0.0082

0.5246	0.1361	8.0863	-1.6966	0.0123	-0.5319	-0.0078
0.6319	0.1212	6.2858	-1.4335	0.0102	-0.4494	-0.0068
0.7464	0.0849	4.2463	-1.0668	0.0075	-0.3344	-0.0055
0.8688	0.0436	2.2735	-0.6020	0.0043	-0.1887	-0.0035
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
PPH+2-BOE						
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0996	0.0646	2.3489	-0.6828	0.0001	-0.2141	-0.0054
0.1994	0.1076	4.0629	-1.1706	0.0001	-0.3670	-0.0077
0.2992	0.1337	5.6540	-1.5262	0.0005	-0.4785	-0.0092
0.3990	0.1500	6.6338	-1.7117	0.0008	-0.5366	-0.0098
0.4990	0.1512	6.9409	-1.7386	0.0009	-0.5451	-0.0098
0.5990	0.1430	6.3881	-1.6026	0.0007	-0.5024	-0.0094
0.6992	0.1221	4.9813	-1.3197	0.0001	-0.4137	-0.0085
0.7994	0.0901	3.2059	-0.9377	-0.0004	-0.2940	-0.0070
0.8996	0.0483	1.3106	-0.4806	-0.0007	-0.1507	-0.0050
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000

TABLE-03c-At T=313.15K

X1	V^E ($\text{cm}^3.\text{mol}^{-1}$)	Δu (m.s^{-1})	Δks (pa-1)	Δz ($10^{-3}/\text{Kg.m}^{-2}.\text{s}^{-1}$)	L_f^E (10-10m)	$\Delta \eta$ (m.pas)
PPH+2-MOE 313.15k						
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0624	0.0127	1.8909	-0.3347	0.0040	-0.1049	-0.0036
0.1302	0.0315	3.7335	-0.6307	0.0076	-0.1977	-0.0051
0.2041	0.0469	5.7482	-0.9095	0.0111	-0.2851	-0.0066
0.2852	0.0596	7.3738	-1.1190	0.0138	-0.3508	-0.0076
0.3744	0.0706	7.9757	-1.2042	0.0150	-0.3775	-0.0078
0.4731	0.0739	7.8447	-1.1894	0.0150	-0.3729	-0.0074
0.5827	0.0673	6.6900	-1.0481	0.0134	-0.3286	-0.0068
0.7054	0.0529	4.8023	-0.7955	0.0103	-0.2494	-0.0056
0.8434	0.0292	2.3618	-0.4360	0.0057	-0.1367	-0.0038
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
PPH+2-EOE						
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0756	0.0309	1.6046	-0.5517	0.0030	-0.1730	-0.0053
0.1554	0.0533	4.0722	-1.1121	0.0066	-0.3486	-0.0073
0.2397	0.0753	5.8213	-1.5093	0.0093	-0.4732	-0.0088
0.3291	0.0901	6.7757	-1.7428	0.0109	-0.5464	-0.0095
0.4239	0.0982	6.9354	-1.8138	0.0114	-0.5686	-0.0097
0.5246	0.0975	6.3764	-1.7298	0.0109	-0.5423	-0.0093
0.6319	0.0866	5.1863	-1.4965	0.0094	-0.4691	-0.0085
0.7464	0.0658	3.0760	-1.0862	0.0065	-0.3405	-0.0075
0.8688	0.0373	1.3650	-0.5944	0.0034	-0.1864	-0.0055
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
PPH+2-BOE						
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0996	0.0459	1.8441	-0.6319	-0.0002	-0.1981	-0.0077
0.1994	0.0721	3.4592	-1.1158	-0.0001	-0.3498	-0.0099
0.2992	0.0922	4.8040	-1.4519	0.0002	-0.4552	-0.0113
0.3990	0.1035	5.7821	-1.6430	0.0006	-0.5151	-0.0114
0.4990	0.1055	6.0839	-1.6727	0.0007	-0.5244	-0.0113
0.5990	0.1016	5.4121	-1.5277	0.0003	-0.4789	-0.0110
0.6992	0.0932	4.2667	-1.2635	-0.0002	-0.3961	-0.0108
0.7994	0.0709	2.6163	-0.8901	-0.0007	-0.2790	-0.0091
0.8996	0.0423	0.7783	-0.4363	-0.0011	-0.1368	-0.0070
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000

TABLE-03d-At T=318.15K

X1	V^E ($\text{cm}^3.\text{mol}^{-1}$)	Δu (m.s^{-1})	Δks (pa-1)	Δz ($10^{-3}/\text{Kg.m}^{-2}.\text{s}^{-1}$)	L_f^E (10-10m)	$\Delta \eta$ (m.pas)
PPH+2-MOE 318.15K						
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0624	0.0067	1.5469	-0.3004	0.0039	-0.0942	-0.0048
0.1302	0.0216	3.1767	-0.5743	0.0074	-0.1800	-0.0066
0.2041	0.0337	4.9333	-0.8296	0.0106	-0.2601	-0.0079
0.2852	0.0486	5.9611	-0.9858	0.0127	-0.3090	-0.0088
0.3744	0.0590	6.1740	-1.0386	0.0136	-0.3256	-0.0090
0.4731	0.0645	5.7891	-1.0033	0.0133	-0.3145	-0.0087

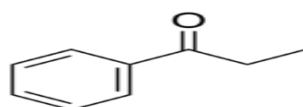
0.5827	0.0579	4.6753	-0.8679	0.0117	-0.2721	-0.0079
0.7054	0.0425	3.0047	-0.6376	0.0088	-0.1999	-0.0069
0.8434	0.0194	1.2931	-0.3429	0.0048	-0.1075	-0.0049
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
PPH+2-EOE						
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0756	0.0151	1.1673	-0.5923	0.0027	-0.1857	-0.0069
0.1554	0.0353	3.1559	-1.1765	0.0058	-0.3688	-0.0092
0.2397	0.0541	4.7490	-1.6147	0.0082	-0.5062	-0.0106
0.3291	0.0750	5.7414	-1.8833	0.0097	-0.5904	-0.0109
0.4239	0.0871	5.9810	-1.9737	0.0103	-0.6188	-0.0109
0.5246	0.0888	5.5437	-1.8935	0.0098	-0.5936	-0.0104
0.6319	0.0797	4.4677	-1.6429	0.0084	-0.5151	-0.0097
0.7464	0.0544	2.5209	-1.1996	0.0058	-0.3761	-0.0089
0.8688	0.0268	0.8897	-0.6479	0.0029	-0.2031	-0.0069
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
PPH+2-BOE						
0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
0.0996	0.0278	1.3106	-0.5886	-0.0005	-0.1845	-0.0102
0.1994	0.0507	2.7175	-1.0612	-0.0005	-0.3327	-0.0126
0.2992	0.0728	4.1243	-1.4115	-0.0001	-0.4425	-0.0140
0.3990	0.0897	4.8584	-1.5830	0.0000	-0.4963	-0.0141
0.4990	0.0947	5.0134	-1.6024	-0.0001	-0.5024	-0.0138
0.5990	0.0908	4.5221	-1.4744	-0.0003	-0.4622	-0.0136
0.6992	0.0813	3.3609	-1.2074	-0.0008	-0.3785	-0.0131
0.7994	0.0570	1.9219	-0.8483	-0.0012	-0.2660	-0.0114
0.8996	0.0289	0.3922	-0.4136	-0.0013	-0.1297	-0.0086
1.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000

3.1 Excess molar volume (V_m^E)

Fig 01 and Table 03 illustrates that the excess molar volumes for all the three binary systems over the whole composition range are Positive and the value decreases with increase in temperatures at 303.15K, 308.15K, 313.15K, 318.15K. The maximum deviation in excess molar volume is obtained at 0.4730, 0.4239 and 0.4989 mole fractions of PPh for (PPh+2-MOE), PPh+2-EOE) and (PPh+2-BOE) respectively. The Excess molar volume (V_m^E) values depends upon three factors mainly physical, chemical and structural properties. The sign and magnitude of the excess properties depend on the following factors [7]:

- (i) The disruption of dipolar molecules in the like associated molecules is due to the addition of second component molecule.
- (ii) Chemical association between unlike molecules through the formation of hydrogen bond, which is a strong specific interaction,
- (iii) Association through weaker physical forces like dipole-dipole interaction and
- (iv) Accommodation of molecules of one component in to the interstitial positions of the structural network of molecules of other component.

The first factor contributes to the positive excess molar volume (V_m^E) due to the expansion in volume and the other three contributes negative excess molar volume. In our present binary systems shows positive excess molar volumes. The addition of propiophenone to alkoxy ethanols, first breaks the intra molecular hydrogen bond within alkoxy ethanols, and then forms a new hydrogen bond between hetero molecules leading to a complex formation. It is assumed that in the present case there is a intermolecular hydrogen bonding is expected than that of intramolecular hydrogen bonding because of polarized C=O.....H bond it causing the breakage of self-association. This is evident from the IR graphs which are discussed in separate section of this paper. The observed higher values of PPh+2-BOE indicate the dominance of molecular dissociation over association. In addition to this the interstitial accommodation of MOE, EOE and BOE molecules into the voids of PPH, the molar volume of PPH ($133.5789 \text{ cm}^3 \text{ mol}^{-1}$) is greater than the two alkoxy-ethanols under study at 303.15 K. ($V_m = 79.8950 \text{ cm}^3 \text{ mol}^{-1}$ for MOE, $97.8321 \text{ cm}^3 \text{ mol}^{-1}$ for EOE and $132.4226 \text{ cm}^3 \text{ mol}^{-1}$ for BOE) is expected, but alkoxy ethanol molecules being expected to exist in rings of 5/6/7 members, it is expected that the molar volume remains positive even though there is possibility of hydrogen bond formation between the C=O group of Propiophenone and the -OH group of alkoxyethanol, the same is observed in the research analysis of anisaldehyde with alkoxyethanol done by Zereena [9] and indicating a very possible reason for the positive values of V_m^E . Same is observed in the research analysis of anisaldehyde with cresols by Narendra et al. [16].



In alkoxyalcohols, the hydrogen atom of alcohols forms hydrogen bond with C=O of the Propiophenone. The newly formed inter-molecular hydrogen bond has more effect than the intramolecular Hydrogen bonding causing the breakage of self-associates. The observed higher positive values of (V_m^E) over the entire range of mole fraction in PPH+BOE system may be attributed to the dominance of molecular dissociation over association. In all systems the V_m^E values decreased with a rise in temperature from 303.15 to 318.15 K. This may be attributed to the rise in kinetic energy of interacting molecules and breakage of intermolecular association followed by new hydrogen bond formations. The algebraic values of (V_m^E) for the studied systems are in the order:

MOE < EOE < BOE:

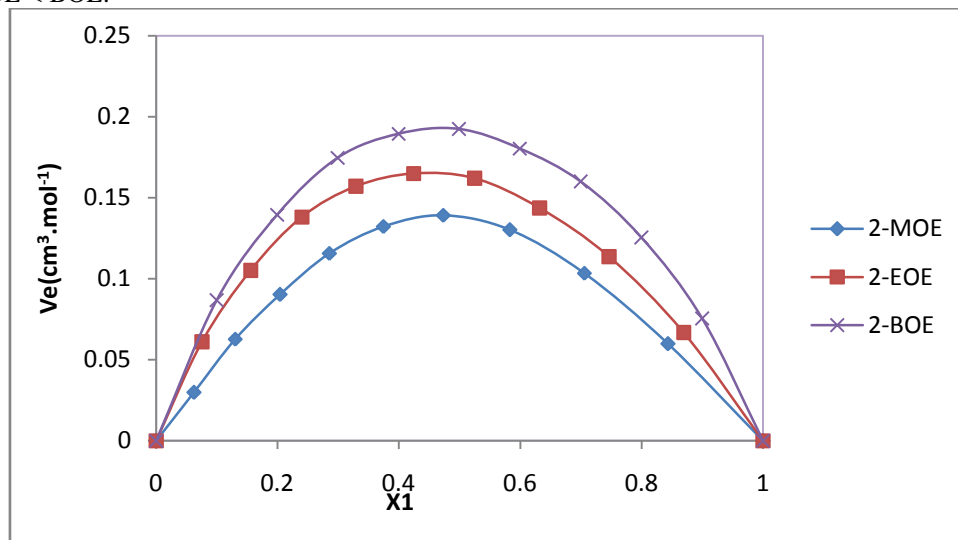


Figure-01: Variation of excess molar Volume with mole fraction of PPH with 2-MOE, 2-EOE & 2-BOE at the Temperature 303.15k

3.2 Deviations in isentropic compressibility and other derived parameters (ΔK_s)

Isentropic compressibility, K_s and deviation in isentropic compressibility ΔK_s , were calculated using the following relations:

$$K_s = \frac{1}{\rho^2} \left(\frac{\partial \rho}{\partial p} \right)_s$$

$$K_s^E = K_s - K_s^{iD}$$

$$K_s^{iD} = \sum_{i=1}^2 \phi_i \left\{ K_{si} + \frac{TV_i(\alpha_i^2)}{C_{pi}} \right\} - \left\{ T \left(\sum_{i=1}^2 x_i V_i \right) \left(\sum_{i=1}^2 \phi_i \alpha_i \right)^2 / \left(\sum_{i=1}^2 x_i C_{pi} \right) \right\}$$

Where u and K_s are the speed of sound, isentropic compressibility of the mixture and K_{si} , the isentropic compressibility of the i^{th} component in the mixture, respectively. The experimental speed of sound, isentropic compressibility and deviation in isentropic compressibility are listed in Table 03 and are graphically represented in Fig 02 as a function of the mole fraction of Propiophenone

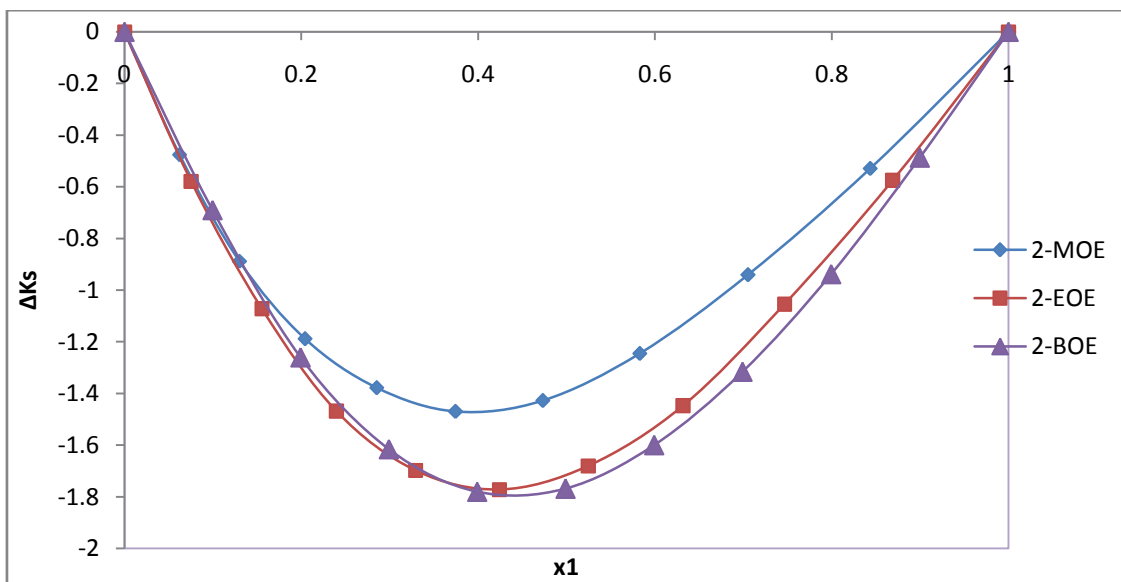


Figure-02: Deviation in isentropic compressibility with mole fraction of PPH with 2-MOE, 2-EOE & 2-BOE at the Temperature 303.15k

Fig:02 shows that ΔK_s values are negative for all the mixtures, this trend is justified by the presence of weak interaction or structure disruptive effects between the mixing liquids for the binary mixtures of alkoxyethanols. The graphical variations of K_s^E values in Fig: 02 as a function of PPH mole fraction (x_1) are negative over the whole composition range for all binary systems investigated. The values of K_s^E become more negative as the number of $-CH_2-$ units in the alkoxyethanol molecules increases (the more negative K_s^E values have been found for the BOE & EOE mixtures). The variation of K_s^E with the size of the alkoxyethanol is consistent with that of the excess molar volume (V_s^E). In general, the values of K_s^E can be considered as arising from two types of interaction between the component molecules:

- (i) a physical interaction, consisting of dispersion forces or weak dipole-dipole interaction making a positive contribution,
- (ii) a chemical or specific interaction, which include charge-transfer forces, forming H-bonds and other complex forming interactions, resulting in a negative contribution to the K_s^E value.

As stated earlier alkoxyethanols in pure state associate predominantly to form ten-membered dimeric rings. These structures can thus resist the structure disruptions in the presence of Propiophenone and this effect probably increases with the number of carbon atoms in the alkoxyethanols. Similar type of results was reported earlier by Sastry and Patel [17].

In an attempt to explore the nature of the interactions, various thermodynamic parameters like intermolecular free length, L_f [18]; specific acoustic impedance, Z [19]; of the binary mixtures have been calculated using the following equations:

$$L_f = K \rho^{\frac{1}{2}} \dots \dots \dots (8)$$

$$Z = u \rho \dots \dots \dots (9)$$

ΔL_f is negative for all the binary mixtures at different temperatures are observed. ΔZ is positive for MOE & EOE mixtures, whereas it is positive at lower temperatures and completely negative at higher temperature for BOE binary mixture. Positive and negative deviations in these functions from rectilinear dependence on composition of the mixtures indicate the extent of association or dissociation between the mixing components [20]. Thus the graded trend obtained from these results supports our earlier results. Hence, negative values of excess isentropic compressibility and excess intermolecular free lengths in the present system suggest that strong intermolecular interactions are present between unlike molecules in the liquid mixtures.

3.3 Excess intermolecular free length (L_f^E)

Ultrasound waves are high frequency mechanical waves. Their velocities in a medium depend inversely on the density and compressibility of the medium. The variation of ultrasonic velocity in a mixture depends upon the increase or decrease of intermolecular free length (L_f), after mixing the components computed excess intermolecular free length L_f^E and is graphically represented in Fig-03.

$$L_f^E = L_f - (x_1 L_{f1} + x_2 L_{f2})$$

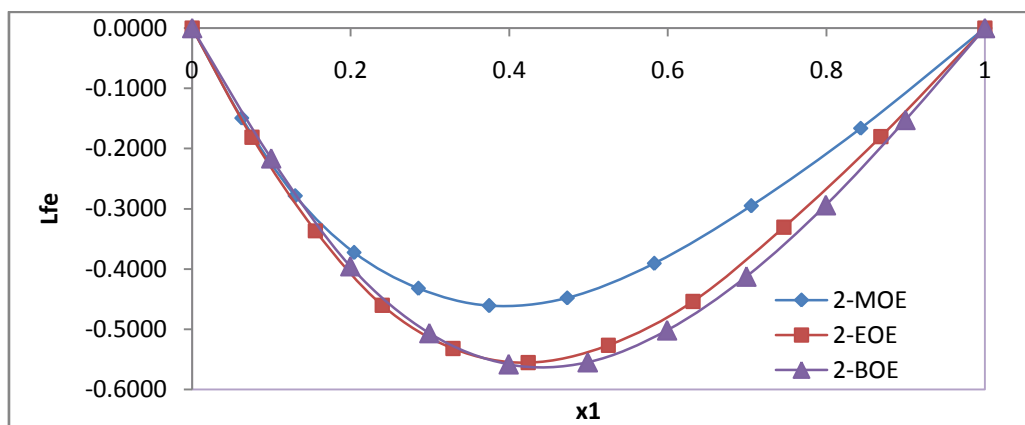


Figure-03: Variation of excess intermolecular free length with mole fraction of PPH with 2-MOE, 2-EOE & 2-BOE at the Temperature 303.15k

Generally, negative values of Δu indicate dispersion forces due to weak interactions whereas positive values of Δu indicate strong interactions [21]. The sign and magnitude of Δu play important roles in describing molecular rearrangements among the component molecules in the mixtures which reflect intermolecular interactions between the molecules. Table 03 shows that the values of Δu are positive for all the binary mixtures of PPH with MOE, EOE & BOE.

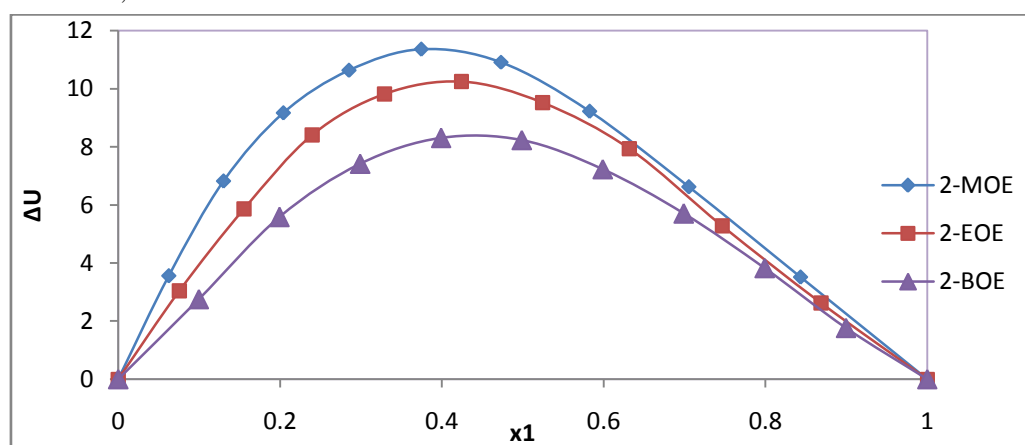


Figure-04: Deviation in ultrasonic speed velocity with mole fraction of PPH with 2-MOE, 2-EOE & 2-BOE at the Temperature 303.15k

An examination of data in Table 03 shows that the excess isentropic compressibility (K_s^E) and excess intermolecular free length (L_f^E) in Table 03 are negative in all of the binary systems over the entire range of composition. According to Sri Devi et al. [22], negative excess values are due to closely packed molecules which accounts for the existence of strong molecular interactions, whereas positive excess values reflect weak interactions between unlike molecules. The sign of the excess isentropic compressibility (K_s^E) and excess intermolecular free length (L_f^E) are useful in assessing the compaction due to molecular interactions in liquid mixtures through: hydrogen-bonding, charge transfer, dipole-dipole and dipole-induced dipole interactions, interstitial accommodation and orientation ordering [23], which lead to a more compact structure, leading to negative values of the excess isentropic compressibility and excess intermolecular free length. Hence negative values of the excess isentropic compressibility (K_s^E) and excess intermolecular free length (L_f^E) in the present systems suggests that strong molecular interactions are present between unlike molecules in the liquid mixtures.

3.4 Deviation in Viscosity ($\Delta\eta$)

The deviation in Viscosity is calculated as below for the experimental data of viscosity (η) of the binary mixtures.

$$\Delta\eta = \eta - \sum_{i=1}^2 (x_i \eta_i)$$

where η and η_i are viscosities of the mixture and the pure compounds respectively.

The nature of intermolecular forces' attraction between the molecules of the binary liquid mixtures can be analyzed by using the viscosity measurements. The values of $\Delta\eta$ are negative for all the binary mixtures of PPH+2MOE, PPH+2EOE, and PPH+2BOE at the temperatures 303.15 to 318.15. In Table-03a to 3d, the viscosity deviation may be generally explained by considering the following factors [24].

- (i) The difference in size and shape of the component molecules and the loss of dipolar association in pure component may contribute to a decrease in viscosity and
- (ii) Specific interactions between unlike components such as hydrogen bond formation and charge-transfer complexes may cause increase in viscosity in mixtures compared to pure components. The former effect produces negative deviation in viscosity and latter effect produces positive viscosity deviation.

This further supports the molecular association through hydrogen bonding between unlike molecules as suggested earlier by other workers [25]. The trend is in negative deviation of viscosity $\Delta\eta$ is MOE>EOE>BOE.

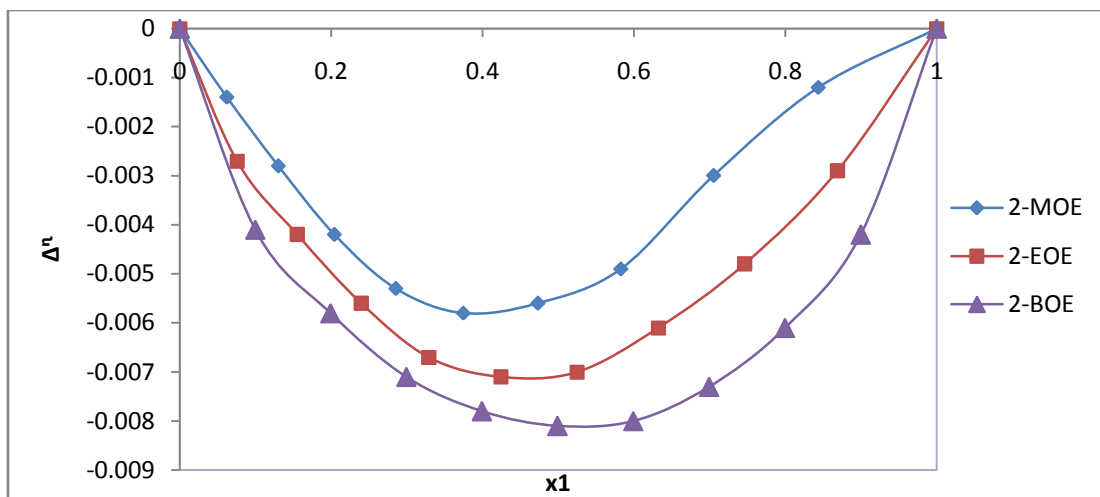


Figure-05: Deviation of Viscosity with mole fraction of PPH with 2-MOE, 2-EOE & 2-BOE at the Temperature 303.15k

The variation of $V^E K_s^E \Delta\eta$ and Δu with mole fraction were fitted to the Redlich–Kister Equation of the type:

$$Y^E = x_1 x_2 \{a_0 + a_1(x_1 - x_2) + a_2(x_1 - x_2)^2\} \dots \dots \dots (10)$$

Where Y^E is $V^E, \Delta u, K_s^E$ or $\Delta\eta$. The values of a_0, a_1 and a_2 are the coefficients of the polynomial equation and were obtained by the method of least-squares and are given in Table 04 along with standard deviation values. The standard deviations are calculated by using the equation:

$$\sigma(Y^E) = \frac{\{\sum_{i=1}^n (Y_{obs}^E - Y_{cal}^E)^2\}^{1/2}}{n-m} \dots \dots \dots (11)$$

where n is the total number of experimental points and m is the number of coefficients.

TABLE- 04– Coefficients of Redlich-Kister Polynomial Equation

Property	Temp(K)	PPH+2-MOE				PPH+2-EOE				PPH+2-BOE			
		A0	A1	A2	σ	A0	A1	A2	σ	A0	A1	A2	σ
VE	303.15	0.5553	-0.0720	-0.1262	0.0012	0.6499	-0.1590	0.1071	0.0012	0.7629	0.0818	0.2077	0.0013
	308.15	0.3844	-0.0383	-0.0665	0.0017	0.5566	-0.1423	-0.1285	0.0016	0.6062	0.1019	0.0326	0.0019
	313.15	0.2970	-0.0394	-0.1210	0.0018	0.3889	-0.0656	-0.0230	0.0013	0.4217	0.0168	0.0993	0.0017
	318.15	0.2570	-0.0326	-0.2000	0.0014	0.3564	-0.0141	-0.2120	0.0013	0.3800	0.0195	-0.1051	0.0018
Δu	303.15	42.3929	23.3428	-0.9854	0.1244	39.6365	15.6274	11.6714	0.2000	33.0661	8.3002	11.8779	0.1680
	308.15	37.4089	17.2192	12.9309	0.1323	33.6790	14.5509	-9.5454	0.3563	27.3701	5.9161	11.4982	0.1721
	313.15	31.2364	12.6245	12.1711	0.2372	27.4654	12.0388	15.5020	0.3423	24.0520	5.9190	14.6701	0.1753
	318.15	23.2450	14.5116	10.2179	0.2793	23.8268	10.1532	18.1780	0.2635	20.3029	5.4524	16.6864	0.1092
Δk_s	303.15	-5.5709	2.6305	-0.4581	0.0070	-6.9140	2.2361	0.5929	0.0180	-7.0957	1.5842	0.8114	0.0186
	308.15	-5.2438	2.1321	0.5952	0.0112	-6.9702	2.2676	0.3584	0.0307	-6.9132	1.3424	0.7517	0.0118
	313.15	-4.7135	1.7419	0.6238	0.0185	-7.1819	2.1579	1.0214	0.0360	-6.6585	1.2936	1.1222	0.0103
	318.15	-3.9918	1.8595	0.4657	0.0222	-7.8285	2.1946	1.2490	0.0312	-6.4291	1.2066	1.3236	0.0059
Δz	303.15	0.0658	-0.0277	0.0020	0.0001	0.0557	-0.0154	-0.0127	0.0002	0.0075	0.0061	-0.0137	0.0002
	308.15	0.0636	-0.0229	-0.0107	0.0002	0.0505	-0.0145	-0.0082	0.0004	0.0033	0.0041	-0.0108	0.0002
	313.15	0.0596	-0.0188	-0.0095	0.0002	0.0457	-0.0131	-0.0145	0.0003	0.0026	0.0151	-0.0151	0.0002
	318.15	0.0528	-0.0214	-0.0054	0.0002	0.0410	-0.0121	-0.0149	0.0002	0.0000	0.0049	-0.0151	0.0001
$\Delta \eta$	303.15	-0.0223	0.0122	0.0114	0.0001	-0.0274	0.0062	-0.0053	0.0002	-0.0314	0.0009	-0.0213	0.0002
	308.15	-0.0259	0.0117	-0.0030	0.0002	-0.0308	0.0106	-0.0148	0.0002	-0.0380	0.0034	-0.0287	0.0003
	313.15	-0.0279	0.0126	-0.0227	0.0004	-0.0358	0.0097	-0.0376	0.0004	-0.0435	0.0045	-0.0561	0.0005
	318.15	-0.0314	0.0150	-0.0389	0.0006	-0.0395	0.0127	-0.0583	0.0006	-0.0529	0.0089	-0.0758	0.0007

3.5 FT IR SPECTRAL STUDY

FT IR Spectroscopy is a useful tool to study the inter and intra molecular hydrogen bonding between the components. In the present study PPh +2-Alkoxy alkanols the absorption band appears around (3160-3640)cm⁻¹[9] due to free –OH group in pure state .The 2-alkoxy alkanols namely 2-Methoxy Ethanol(MOE),2-EthoxyEthanol(EOE),2-ButoxyEthanol(BOE) exhibits some characteristic absorption peaks at 3396cm⁻¹(O-Hstretch), 3412cm⁻¹(O-H stretch),3402cm⁻¹(O-Hstretch)and in pure propiophenone no O-H Stretch absorptions, only C=OStretch at 1681cm⁻¹ respectively. In the present investigation FT-IR spectrum considered at room temperature and 1:1 ratio, the resultant absorption bands of (PPh+2-MOE),(PPh+2-EOE)and (PPh+2-BOE) are 3429cm⁻¹,3431cm⁻¹ and3448cm⁻¹ respectively, this indicates that frequency shift increases from MOE to BOE, it is known that for less extensive hydrogen bonding a sharper and less intense band at higher frequency but due to extensive hydrogen bonding ,a broad band appears at a lower frequency. In case of BOE showing weak inter molecular interactions that causes change in the frequency shift in hydrogen bond intensity .This indicates that lesser strength of association between BOE as compared to other alkoxy ethanols, even at higher concentrations the formation of intermolecular hydrogen bonding is lesser but showing self-association because of steric hindrance by longer carbon chains, dispersive forces by the monomers, longer intermolecular free lengths and lesser ultrasonic sound velocities.

Shift in hydrogen bond intensity –OH(ν cm⁻¹): comparison of all three 2-Alkoxy alkanols

Pure/Binarymixture	MOE	EOE	BOE
Pure	3396cm ⁻¹	3412cm ⁻¹	3402 cm ⁻¹
0.25M+0.75M	3439 cm ⁻¹	3402 cm ⁻¹	3419 cm ⁻¹
0.5M+0.5M	3429 cm ⁻¹	3431 cm ⁻¹	3448 cm ⁻¹
0.75M+0.25M	3450 cm ⁻¹	3481 cm ⁻¹	3487 cm ⁻¹

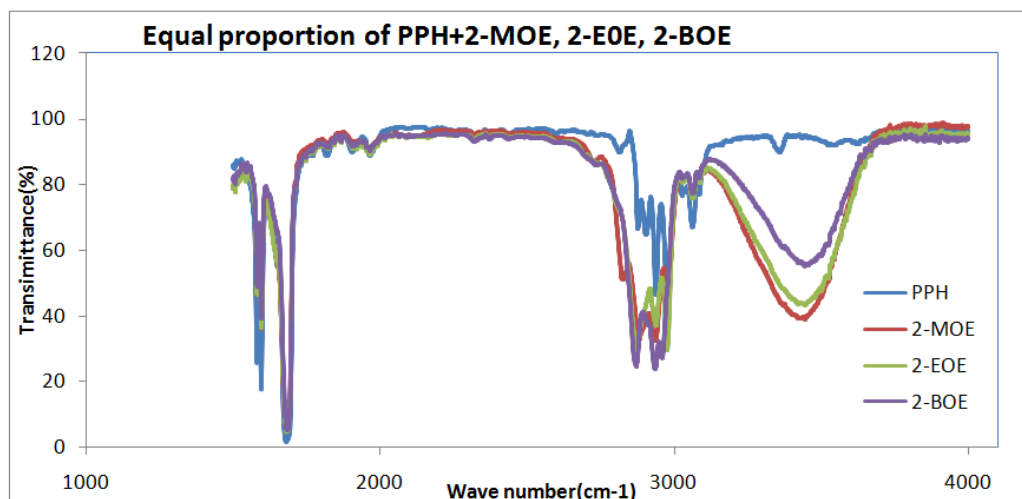


Figure-06: IR spectral study on equal proportion of PPH+2MOE, 2-EOE & 2-BOE

IV. Conclusions

In this paper, the densities, excess molar volume, viscosities and speed of sound at temperatures 303.15K, 308.15K, 313.5K and 318.15K over the entire range of composition of Propiophenone with alkoxyethanols have been measured. From these measured physical property data, excess molar volumes, deviation in viscosity, excess ultrasonic velocity and excess isentropic compressibility have been calculated and correlated by a Redlich–Kister type polynomial equation to derive the coefficients and standard deviation. The intermolecular interactions of the components are interpreted and found the results as positive and negative for excess molar volume and viscosity respectively. It is observed that an inter and intramolecular association is formed between Propiophenone and 2- alkoxyethanols. FT IR data indicates the strength of inter and intra molecular association between carbonyl group of Propiophenone with OH group of 2-Alkoxy alkanols.

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